

This worksheet calculates the transient advection, dispersion, and decay of tritium in the vadose zone. Tritiated water is transported in both the aqueous and vapor phases. Local thermodynamic equilibrium is assumed so that the partitioning of tritium between the aqueous and gas phases can be expressed using Henry's Law. Tritium undergoes radioactive decay with a half-life of 12.3 years. This solution is derived in Jury et al., 1983, J. Env. Qual., 12, 558-564 and in Jury et al., 1990, WRR, 26(1), 13-20.

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### **Input parameters and distributions:**

Define number of realizations:  $n := 100$

$i := 1 .. n$

Total porosity:  $\phi := \text{runif}(n, .302, .445)$   $\text{median}(\phi) = 0.378$

Moisture Content:  $\theta_w := \text{runif}(n, .053, .225)$

Gas Content:  $a := \phi - \theta_w$   $\text{max}(a) = 0.38$

Henry's Constant:  $K_H := 17 \cdot 10^{-6}$

Adsorption partition coefficient:  $K_d := 0 \cdot \frac{m^3}{kg}$

Liquid-Phase Diffusion Coefficient:  $D_w := 2.26 \cdot 10^{-9} \cdot \frac{m^2}{s}$  See Smiles et al. 1995

Gas-Phase Diffusion Coefficient:  $D_g := 2.57 \cdot 10^{-5} \cdot \frac{m^2}{s}$

Half-life:  $t_{\text{half}} := 12.3 \cdot 365.25 \cdot 24 \cdot 3600 \cdot s$

Bulk density:  $\rho_b := \text{runif}(n, 1.47, 1.85) \cdot kg \cdot m^{-3}$   $\text{median}(\rho_b) = 1.621 \text{ kg m}^{-3}$

Inventory (Ci/Specific Activity/1000 g/kg):  $C_i := \text{runif}(n, 2400, 4800)$   
Estimated initial inventory = 2400 Ci  $\text{mean}(C_i) = 3.556 \times 10^3$

Specific Activity (Ci/g):  $SA := 9700$

Waste zone length:  $\text{length} := \text{runif}(n, 10 \cdot .3048, 430 \cdot .3048) \cdot \text{m}$  mean(length) = 73.205 m

Waste zone width:  $\text{width} := \text{runif}(n, 10 \cdot .3048, 300 \cdot .3048) \cdot \text{m}$  mean(width) = 48.092 m

Minimum length and width is determined by size of pit 33 (10'x10')

Maximum length and width is determined by extent of MWL.

Waste zone thickness:  $L := \text{runif}(n, 10 \cdot .3048, 27 \cdot .3048) \cdot \text{m}$  mean(L) = 5.697 m  
 $L = 22 \text{ ft}$

Distance to water table:  $d_{\text{wt}} := \text{runif}(n, 461 \cdot .3048, 495 \cdot .3048) \cdot \text{m}$

Thickness of clean overburden:  $L_c := \text{runif}(n, 0 \cdot .3048, 16 \cdot .3048) \cdot \text{m}$   
6 to 16 feet nominal; 0 minimum is due to erosion

Darcy Infiltration:  $q := \text{runif}(n, 1.18 \cdot 10^{-11}, 6.12 \cdot 10^{-11}) \cdot \frac{\text{m}}{\text{s}}$

Surface boundary-layer thickness:  $d_{\text{BL}} := \text{runif}(n, 0.001, 1) \cdot \text{m}$

Tortuosity Factor:  $\varepsilon_w := \text{runif}(n, 0.001, 1)$

(lower bound from Millington 1959; upper bound is physical limit)  $\varepsilon_g := \text{runif}(n, 0.1, 1)$

Tritiated water inhalation dose conversion factor (rem/pCi):  $\text{DCF}_{\text{tritium}} := 6.4 \cdot 10^{-11}$

US Environmental Protection Agency. 1988. Federal guidance report no. 11: limiting values of radionuclide intake and air concentration and dose conversion factors for inhalation, submersion, and ingestion, Eckerman, K.F., A.B. Wolbarst, and A.C.B. Richardson, Washington, DC: US Environmental Protection Agency. Report No.: EPA-5201/1-88-020.

### Input Parameter Distributions for sensitivity analyses:

$$A_i := \text{length}_i \cdot \text{width}_i$$

	1
1	$1.557 \cdot 10^3$
2	$5.514 \cdot 10^3$
3	$5.14 \cdot 10^3$

$$\text{m}^2$$

	1
1	0.128
2	0.152
3	0.161
4	0.14

	1
1	$4.14 \cdot 10^3$
2	$4.066 \cdot 10^3$
3	$3.972 \cdot 10^3$

	1
1	0.773
2	0.321
3	0.146

	1
1	0.637
2	0.885
3	0.313
4	0.311

	1
1	0.364
2	4.461
3	3.684

	1
1	146.778
2	144.257
3	150.627

	1
1	$1.915 \cdot 10^{-11}$
2	$1.731 \cdot 10^{-11}$

$$q = \begin{array}{|c|c|} \hline & 1 \\ \hline 1 & 1.915 \cdot 10^{-11} \\ \hline 2 & 1.731 \cdot 10^{-11} \\ \hline \end{array} \text{ m s}^{-1} \quad \phi = \begin{array}{|c|c|} \hline & 1 \\ \hline 1 & 0.302 \\ \hline 2 & 0.33 \\ \hline 3 & 0.386 \\ \hline \end{array} \quad d_{BL} = \begin{array}{|c|c|} \hline & 1 \\ \hline 1 & 0.05 \\ \hline 2 & 0.118 \\ \hline 3 & 0.74 \\ \hline 4 & 5.411 \cdot 10^{-3} \\ \hline 5 & 0.956 \\ \hline \end{array} \text{ m} \quad L = \begin{array}{|c|c|} \hline & 1 \\ \hline 1 & 5.184 \\ \hline 2 & 6.162 \\ \hline 3 & 5.513 \\ \hline 4 & 7.977 \\ \hline \end{array} \text{ m}$$

### Calculated Parameters

Initial mass in kilograms:

$$M_{o_i} := \frac{C_i}{SA \cdot 1000} \cdot kg$$

Volume of waste zone:

$$V_i := \text{length}_i \cdot \text{width}_i \cdot L_i$$

Source Concentration (Total):

$$C_{o_i} := \frac{M_{o_i}}{V_i}$$

Retardation factor:

$$R_{L_i} := \rho_{b_i} \cdot K_d + \theta_{w_i} + a_i \cdot K_H$$

Effective Diffusion Coefficient:

$$D_{E_i} := \frac{\left( \varepsilon_{w_i} \cdot D_w + \varepsilon_{g_i} \cdot K_H \cdot D_g \right)}{R_{L_i}}$$

Effective Velocity:

$$V_{E_i} := \frac{q_i}{R_{L_i}}$$

Decay constant:

$$\mu := \frac{\ln(2)}{t_{\text{half}}} \quad \mu = 1.786 \times 10^{-9} \text{ s}^{-1}$$

Surface mass-transfer coefficient:

$$H_{E_i} := \frac{D_g}{d_{BL_i}} \cdot \frac{K_H}{R_{L_i}}$$

The  $d_{wt}$  parameter can be assigned a constant value ( $d_{wt,i}$ ) to simulate concentrations at different depths.

$$\text{Note: } R_L / K_H = R_G$$

	1
1	146.778
2	144.257
3	150.627
4	149.796
5	150.394
6	142.603

### Concentration at water table as a function of time:

ntimes := 100

nyears := 1000

$j := 1..n_{\text{times}}$

$n_{\text{sec}} := n_{\text{years}} \cdot 365.25 \cdot 24 \cdot 3600$

$$\text{Time: } t_j := \frac{j}{n_{\text{times}}} \cdot n_{\text{sec}} \cdot s \quad \text{erfc}(x) := 1 - \text{erf}(x)$$

Parameters for system with clean overburden plus source thickness

$$\exp1_{i,j} := \text{if} \left[ \frac{\left[ H_{E_i} \cdot (H_{E_i} + V_{E_i}) \cdot t_j + (H_{E_i} + V_{E_i}) \cdot d_{wt_i} \right]}{D_{E_i}} > 700, 700, \frac{\left[ H_{E_i} \cdot (H_{E_i} + V_{E_i}) \cdot t_j + (H_{E_i} + V_{E_i}) \cdot d_{wt_i} \right]}{D_{E_i}} \right]$$

$$\exp2_i := \text{if} \left[ \frac{H_{E_i} \cdot (L_i + L_{c_i})}{D_{E_i}} > 700, 700, \frac{H_{E_i} \cdot (L_i + L_{c_i})}{D_{E_i}} \right] \quad \exp3_i := \text{if} \left( \frac{V_{E_i} \cdot d_{wt_i}}{D_{E_i}} > 700, 700, \frac{V_{E_i} \cdot d_{wt_i}}{D_{E_i}} \right)$$

$$A1_{i,j} := \text{erfc} \left( \frac{d_{wt_i} - (L_i + L_{c_i}) - V_{E_i} \cdot t_j}{\sqrt{4 \cdot D_{E_i} \cdot t_j}} \right) \quad A2_{i,j} := \text{erfc} \left( \frac{d_{wt_i} - V_{E_i} \cdot t_j}{\sqrt{4 \cdot D_{E_i} \cdot t_j}} \right)$$

$$A3_{i,j} := \text{erfc} \left( \frac{d_{wt_i} + (L_i + L_{c_i}) + V_{E_i} \cdot t_j}{\sqrt{4 \cdot D_{E_i} \cdot t_j}} \right) \quad A4_{i,j} := \text{erfc} \left( \frac{d_{wt_i} + V_{E_i} \cdot t_j}{\sqrt{4 \cdot D_{E_i} \cdot t_j}} \right) \quad A5_{i,j} := \exp(\exp1_{i,j})$$

$$A6_{i,j} := \text{erfc} \left( \frac{d_{wt_i} + (2 \cdot H_{E_i} + V_{E_i}) \cdot t_j}{\sqrt{4 \cdot D_{E_i} \cdot t_j}} \right) \quad A7_{i,j} := \text{erfc} \left( \frac{d_{wt_i} + (L_i + L_{c_i}) + (2 \cdot H_{E_i} + V_{E_i}) \cdot t_j}{\sqrt{4 \cdot D_{E_i} \cdot t_j}} \right)$$

Parameters for clean overburden alone:

$$\exp2c_i := \text{if} \left( \frac{H_{E_i} \cdot L_{c_i}}{D_{E_i}} > 700, 700, \frac{H_{E_i} \cdot L_{c_i}}{D_{E_i}} \right)$$

$$A1c_{i,j} := \text{erfc} \left( \frac{d_{wt_i} - L_{c_i} - V_{E_i} \cdot t_j}{\sqrt{4 \cdot D_{E_i} \cdot t_j}} \right) \quad A3c_{i,j} := \text{erfc} \left( \frac{d_{wt_i} + L_{c_i} + V_{E_i} \cdot t_j}{\sqrt{4 \cdot D_{E_i} \cdot t_j}} \right)$$

$$A7c_{i,j} := \text{erfc} \left( \frac{d_{wt_i} + L_{c_i} + (2 \cdot H_{E_i} + V_{E_i}) \cdot t_j}{\sqrt{4 \cdot D_{E_i} \cdot t_j}} \right)$$

Concentration with just clean overburden:

$$C_{Tc_{i,j}} := 0.5 \cdot C_{o_i} \cdot \exp(-\mu \cdot t_j) \cdot \left[ A1c_{i,j} - A2_{i,j} + \left( 1 + \frac{V_{E_i}}{H_{E_i}} \right) \cdot \exp(\exp 3_i) \cdot (A3c_{i,j} - A4_{i,j}) \dots \right. \\ \left. + \left( 2 + \frac{V_{E_i}}{H_{E_i}} \right) \cdot A5_{i,j} \cdot (A6_{i,j} - \exp(\exp 2_i) \cdot A7_{i,j}) \right]$$

Concentration with clean overburden plus source thickness:

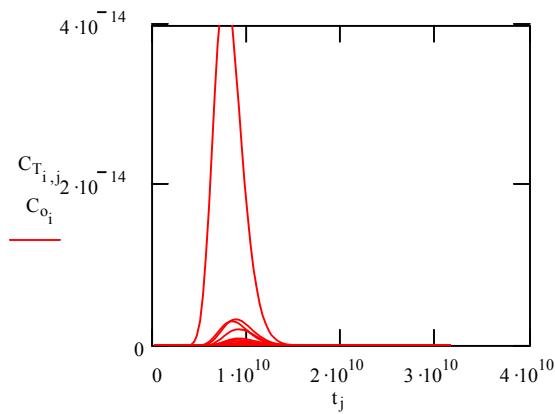
$$C_{Tt_{i,j}} := 0.5 \cdot C_{o_i} \cdot \exp(-\mu \cdot t_j) \cdot \left[ A1_{i,j} - A2_{i,j} + \left( 1 + \frac{V_{E_i}}{H_{E_i}} \right) \cdot \exp(\exp 3_i) \cdot (A3_{i,j} - A4_{i,j}) \dots \right. \\ \left. + \left( 2 + \frac{V_{E_i}}{H_{E_i}} \right) \cdot A5_{i,j} \cdot (A6_{i,j} - \exp(\exp 2_i) \cdot A7_{i,j}) \right]$$

Combined concentration using superposition (Jury et al., 1990, WRR, 26(1), 13-20).

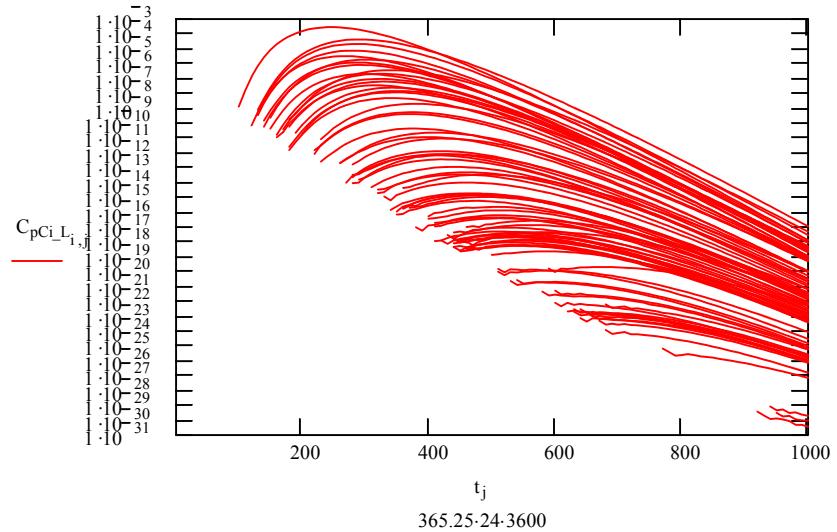
$$C_{T_{i,j}} := C_{Tt_{i,j}} - C_{Tc_{i,j}} \quad \max(C_T) = 1.582 \times 10^{-21} \text{ kg m}^{-3}$$

$$C_{T_{i,j}} := \text{if}(C_{T_{i,j}} < 0, 0, C_{T_{i,j}}) \quad \max(C_{Tt}) = 2.045 \times 10^{-21} \text{ kg m}^{-3}$$

$$\max(C_{Tc}) = 4.624 \times 10^{-22} \text{ kg m}^{-3}$$



$$C_{L_{i,j}} := \frac{C_{T_{i,j}}}{R_{L_i}} \quad C_{pCi\_L_{i,j}} := C_{L_{i,j}} \cdot \frac{1000 \cdot SA \cdot 10^{12}}{1000}$$



$$\max(C_{pCi\_L}) = 2.764 \times 10^{-4} \text{ kg m}^{-3}$$

$$\text{median}(C_{pCi\_L}) = 8.605 \times 10^{-25} \text{ kg m}^{-3}$$

$$\text{mean}(C_{pCi\_L}) = 4.202 \times 10^{-7} \text{ kg m}^{-3}$$

$$C_{pCi\_L} = \text{kg m}^{-3}$$

	1	2	3	4	5
1	0	0	0	0	0
2	0	0	0	0	0
3	0	0	0	0	0
4	0	0	0	0	0
5	0	0	0	0	0
6	0	0	0	0	0
7	0	0	0	0	0

## Calculate surface flux as a function of time

Parameters for source plus clean overburden:

$$J1_{i,j} := \operatorname{erfc}\left(\frac{V_{E_i} \cdot t_j}{\sqrt{4 \cdot D_{E_i} \cdot t_j}}\right) \quad J2_{i,j} := \operatorname{erfc}\left(\frac{L_i + L_{c_i} + V_{E_i} \cdot t_j}{\sqrt{4 \cdot D_{E_i} \cdot t_j}}\right)$$

$$J3_{i,j} := \text{if} \left[ \frac{H_{E_i} \cdot (H_{E_i} + V_{E_i}) \cdot t_j}{D_{E_i}} > 700, 700, \frac{H_{E_i} \cdot (H_{E_i} + V_{E_i}) \cdot t_j}{D_{E_i}} \right]$$

$$J4_i := \text{if} \left[ \frac{H_{E_i} \cdot (L_i + L_{c_i})}{D_{E_i}} > 700, 700, \frac{H_{E_i} \cdot (L_i + L_{c_i})}{D_{E_i}} \right]$$

$$J5_{i,j} := \operatorname{erfc}\left(\frac{(L_i + L_{c_i}) + (2 \cdot H_{E_i} + V_{E_i}) \cdot t_j}{\sqrt{4 \cdot D_{E_i} \cdot t_j}}\right) \quad J6_{i,j} := \operatorname{erfc}\left(\frac{(2 \cdot H_{E_i} + V_{E_i}) \cdot t_j}{\sqrt{4 \cdot D_{E_i} \cdot t_j}}\right)$$

Parameters for clean overburden only:

$$J2_{c_i,j} := \operatorname{erfc}\left(\frac{L_{c_i} + V_{E_i} \cdot t_j}{\sqrt{4 \cdot D_{E_i} \cdot t_j}}\right)$$

$$J4_{c_i} := \text{if} \left[ \frac{H_{E_i} \cdot (L_{c_i})}{D_{E_i}} > 700, 700, \frac{H_{E_i} \cdot (L_{c_i})}{D_{E_i}} \right]$$

$$J5_{c_i,j} := \operatorname{erfc}\left(\frac{(L_{c_i}) + (2 \cdot H_{E_i} + V_{E_i}) \cdot t_j}{\sqrt{4 \cdot D_{E_i} \cdot t_j}}\right)$$

Surface flux with both source and clean overburden:

$$J_{st,i,j} := 0.5 \cdot C_{o_i} \cdot \exp(-\mu \cdot t_j) \cdot [V_{E_i} \cdot (J1_{i,j} - J2_{i,j}) + (2 \cdot H_{E_i} + V_{E_i}) \cdot \exp(J3_{i,j}) \cdot (\exp(J4_i) \cdot J5_{i,j} - J6_{i,j})]$$

Surface flux with just clean overburden:

$$J_{sc,i,j} := 0.5 \cdot C_{o_i} \cdot \exp(-\mu \cdot t_j) \cdot [V_{E_i} \cdot (J1_{i,j} - J2_{c_i,j}) + (2 \cdot H_{E_i} + V_{E_i}) \cdot \exp(J3_{i,j}) \cdot (\exp(J4_{c_i}) \cdot J5_{c_i,j} - J6_{i,j})]$$

Combined concentration using superposition (Jury et al., 1990, WRR, 26(1), 13-20).

$$J_{si,j} := J_{st,i,j} - J_{sc,i,j}$$

This is the downward total flux at the surface. The volatilization flux is equal to the negative of this value.

$$\min(J_{st}) = -1.106 \times 10^{-15} \text{ kg m}^{-2} \text{ s}^{-1}$$

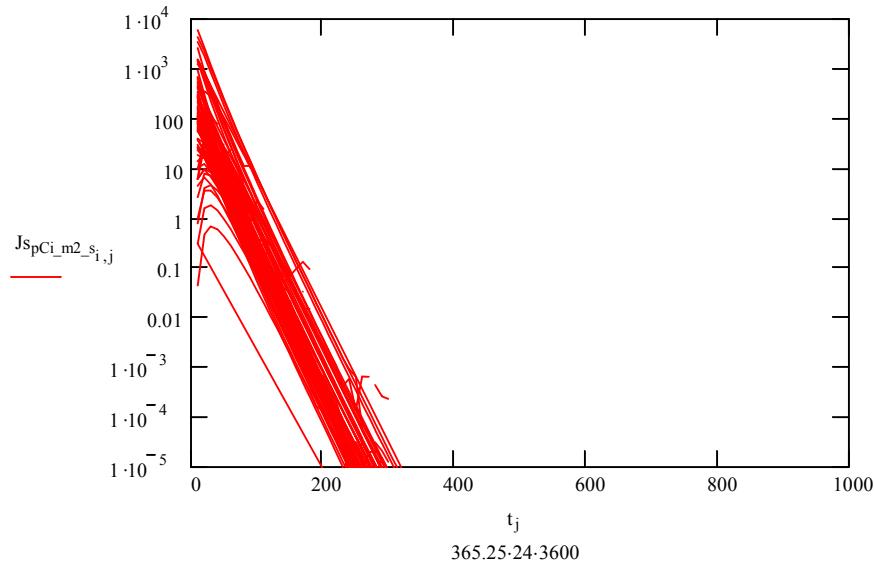
$$\text{mean}(J_{st}) = -1.377 \times 10^{-18} \text{ kg m}^{-2} \text{ s}^{-1}$$

$$\min(J_{sc}) = -6.688 \times 10^{-16} \text{ kg m}^{-2} \text{ s}^{-1}$$

$$\text{mean}(J_{sc}) = -6.954 \times 10^{-19} \text{ kg m}^{-2} \text{ s}^{-1}$$

$$J_{spCi\_m2\_s_{i,j}} := \text{if}[J_{si,j} > 0, 0, -\left(J_{si,j} \cdot 1000 \cdot SA \cdot 10^{12}\right)]$$

This is the surface volatilization flux to the atmosphere in pCi/m^2/s.



$$\max(J_{\text{SpCi\_m2\_s}}) = 5.872 \times 10^3 \text{ kg m}^{-2} \text{ s}^{-1}$$

$$\text{mean}(J_{\text{SpCi\_m2\_s}}) = 6.679 \text{ kg m}^{-2} \text{ s}^{-1}$$

$$J_{\text{SpCi\_m2\_s}} = \begin{array}{|c|c|c|} \hline & 1 & 2 \\ \hline 1 & 0 & 0 \\ \hline 2 & 0 & 0 \\ \hline 3 & 0.984 & 3.566 \\ \hline 4 & 0 & 0 \\ \hline \end{array} \text{ kg m}^{-2} \text{ s}^{-1}$$

**Calculate atmospheric concentration and dose to an individual inhaling the air directly above the waste site.**

Average wind speed (m/s):  $v_{\text{wind}} := 3.63 \cdot \text{m} \cdot \text{s}^{-1}$

(from average of SNL Site Environmental Monitoring Reports 1990-1996)

Vertical atmospheric mixing length (m):  $L_v := 2 \cdot \text{m}$

(conservative value to encompass volume most likely occupied by the average human, Yu, C. et al., 1993, Data Collection Handbook to Support Modeling the Impacts of Radioactive Material in Soil, ANL/EAIS-8, Argonne National Laboratory, Argonne, Illinois.)

$$\text{length} = \begin{array}{|c|c|} \hline & 1 \\ \hline 1 & 20.771 \\ \hline 2 & 73.51 \\ \hline 3 & 62.373 \\ \hline \end{array} \text{ m}$$

$$\text{width} = \begin{array}{|c|c|} \hline & 1 \\ \hline 1 & 74.983 \\ \hline 2 & 75.016 \\ \hline \end{array} \text{ m}$$

Lateral atmospheric mixing distance (m):  $L_{h_i} := \text{if}(length_i < width_i, length_i, width_i)$

(the lateral atmospheric mixing distance is conservatively estimated as the minimum of either the length or width of the waste zone)

Flow rate of air in mixing volume:  $Q_{\text{wind}_i} := v_{\text{wind}} \cdot L_v \cdot L_{h_i}$

Tritium concentration in atmosphere  
(pCi/m<sup>3</sup>):

$$C_{atm\_pCi\_m3} := \frac{J s_{pCi\_m2\_s_{i,j}} \cdot length_i \cdot width_i}{Q_{wind_i}}$$

$$C_{atm\_pCi\_m3} = \begin{array}{|c|c|} \hline & 1 \\ \hline 1 & 0 \\ \hline 2 & 0 \\ \hline 3 & 11.168 \\ \hline \end{array} \text{ kg m}^{-3}$$

Inhalation rate:

$$I := \frac{20}{24 \cdot 3600} \cdot m^3 \cdot s^{-1} \quad I = 2.315 \times 10^{-4} m^3 s^{-1}$$

(20 m<sup>3</sup>/day from U.S. EPA 1991, U.S. Environmental Protection Agency. Human health evaluation manual, supplemental guidance: "Standard default exposure factors". OSWER Directive 9285.6-03.

Receptor Intake:

$$Intake_{i,j} := C_{atm\_pCi\_m3_{i,j}} \cdot I \quad \text{This is the receptor intake in pCi/s}$$

Dose (mrem/year):

$$Dose_{mrem\_yr} := Intake_{i,j} \cdot DCF_{tritium} \cdot 1.5 \cdot 1000 \cdot 3600 \cdot 24 \cdot 365.25$$

(The dose conversion factor, DCF, is in units of rem/pCi; the 1.5 factor is used to account for dermal absorption)

$$Dose_{mrem\_yr} = \begin{array}{|c|c|c|} \hline & 1 & 2 \\ \hline 1 & 0 & 0 \\ \hline 2 & 0 & 0 \\ \hline 3 & 7.832 \cdot 10^{-3} & 0.028 \\ \hline 4 & 0 & 0 \\ \hline \end{array} \text{ kg s}^{-1}$$

$$\max(Dose_{mrem\_yr}) = 18.02 \text{ kg s}^{-1}$$

$$\text{mean}(Dose_{mrem\_yr}) = 0.031 \text{ kg s}^{-1}$$

}

$L =$

	1
1	5.184
2	6.162
3	5.513
4	7.977
5	7.374
6	7.202
7	7.556

m

	1
1	146.778
2	144.257
3	150.627
4	149.796

$d_{wt} =$  m